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**(54) Stretched film of lactic acid-based polymer**

(57) The stretched film of the lactic acid-based polymer in the invention can degrade under natural environment, and additionally has an excellent mechanical strength and durability. By use of a lubricant in combination with an inorganic filler, these properties are further improved and thickness accuracy is enhanced. Consequently, the stretched film of the lactic acid-based polymer in the invention can be widely applied to a pre-paid card and other various film materials, lamination materials and packaging materials. Even though abandoned after use in the natural environment, the stretched film relatively quickly decomposes into carbon dioxide and water and does not accumulate as waste.

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Table 1

	Example 1	Example 2	Example 3	Example 4	Example 5	Example 6
Polymer (100 parts by weight)	A	B	C	D	B	B
Filler (parts by weight) (particle size, $\mu$ m)	barium sulfate 5 0.85	titanium oxide 10 0.21	silica 15 5	calcium cabonate 20 1.5	kaolin 23 3.5	Talc 10 1.65
Stretching Ratio (times) longitudinal transverse	2.5 2.5	3 3	1.5 1.5	3.5 3.5	2 3	2 2
Lubricant (parts by weight)	—	—	—	—	—	—
Stretching Ability	○	○	○	○	○	○
Variation Coefficient of Thickness	1.5	1	2	2.5	2.5	1.5
Tensile Strength (MPa) longitudinal transverse	90 88	105 101	85 86	90 92	90 99	95 93
Folding Endurance (the number of reflections)	1100	1800	1000	1200	1400	1400

Table 1 (continued)

	Example 7	Example 8	Example 9	Example 10	Example 11	Example 12
Polymer (100 parts by weight)	B	B	B	B	B	A
Filler (parts by weight) (particle size, $\mu\text{m}$ )	titanium oxide 15 0.21	titanium oxide 10 0.21	titanium oxide 10 0.3	titanium oxide 10 0.21	titanium oxide 10 0.21	barium sulfate 10 1.5
Stretching Ratio (times) longitudinal transverse	3 3	3 3	3 3	3 3	3 3	2.5 2.5
Lubricant (parts by weight)	—	erucamide 1	montan wax 0.2	montan wax 0.2	calcium hydroxy- stearate 1.5	montan wax 0.2
Stretching Ability	○	○	○	○	○	○
Variation Coefficient of Thickness	0.5	0.45	0.4	0.4	0.45	0.8
Tensile Strength (MPa) longitudinal transverse	110 105	111 108	112 110	115 112	110 105	95 93
Folding Endurance (the number of reflections)	2100	2200	2300	2400	2100	1400

Table 1 (continued)

	Example 13	Example 14	Example 15	Example 16
Polymer (100 parts by weight)	B	C	D	B
Filler (parts by weight) (particle size, $\mu\text{m}$ )	Talc 10 8	silica 10 10	calcium carbonate 10 4	kaolin 10 8
Stretching Ratio (times) longitudinal transverse	2.5 2.5	2.5 2.5	2.5 2.5	2.5 2.5
Lubricant (parts by weight)	montan wax 0.2	montan wax 0.2	montan wax 0.2	montan wax 0.2
Stretching Ability	○	○	○	○
Variation Coefficient of Thickness	0.8	1.0	1.2	1.2
Tensile Strength (MPa) longitudinal transverse	96 94	86 88	92 95	95 99
Folding Endurance (the number of reflections)	1500	1100	1300	1600

## Comparative Examples 1 to 4

To each 100 parts by weight of the lactic acid-based polymer A and B which was obtained in Preparation Examples 1 to 2, respective inorganic filler was individually added in an amount shown in Table 2.

Each mixture was treated by the same procedures as carried out in Example 1 to prepare lactic acid-based polymer compositions. The inorganic fillers used were zinc oxide (EP, manufactured by Mitsui Mining & Smelting Co., Ltd.), magnesium oxide (KYOWAMAG150, manufactured by Kyowa Chemical Industry Co., Ltd.) and titanium oxide (TIPAQUE CR60-2, manufactured by Ishihara Sangyo Co., Ltd.). By using the lactic acid-based polymer compositions thus obtained, stretched films were prepared by the same procedures as Example 1 with stretching magnifications shown in Table 1. The tensile strength and folding endurance of the stretched film obtained were measured by the above method and results are illustrated in Table 2.

## Comparative Examples 5 to 14

To each 100 parts by weight of the lactic acid-based polymer A to D which was obtained in Preparation Examples 1 to 4, respective inorganic filler and lubricant were individually added in an amount shown in Table 2. Each mixture was treated by the same procedures as carried out in Example 1 to prepare lactic acid-based polymer compositions. The inorganic fillers used were titanium oxide (TTO-55 and TIPAUQUE CR60-2, manufactured by Ishihara Sangyo Co., Ltd.), barium sulfate (BARIFINE BF-10 and BARIUM SULFATE precipitated SP, manufactured by Sakai Chemical Industry Co., Ltd.), silica (SYLYSIA470, manufactured by Fuji Silysia Chemical Ltd.), calcium carbonate (SS#30, manufactured by Nitto Funaka Kogyo KK), kaolin (NN KAOLIN CLAY, manufactured by Tsuchiya Kaolin Industry Ltd.) and talc (LMS-300 and NK-48, manufactured by Fuji Talc Industrial Co. Ltd.). By using the lactic acid-based polymer compositions thus obtained, stretched films were prepared by the same procedures as Example 1 with stretching magnifications shown in Table 2. The tensile strength and folding endurance of the stretched film obtained were measured by the above method and results are illustrated in Table 2.

## Comparative Example 15

To the lactic acid-based polymer powder B which was obtained in Preparation Example 2, titanium oxide (TIPAUQUE CR60-2, manufactured by Ishihara Sangyo Co., Ltd.) was added in an amount shown in Table 2. The mixture was melt-extruded with a twin screw kneading extruder at cylinder temperature of 170 to 210°C and pelletized. The pellet thus obtained was dried and heat-treated in an oven. After crystallizing the polymer, the pellet was extruded with a T-die mounted single screw extruder at temperature of 150 to 200°C and cooled on a casting roll at 35°C to obtain an unstretched film having an average thickness of 200 µm. The tensile strength and folding endurance of the unstretched film obtained were measured by the above method and results are illustrated in Table 2.

Table 2 (continued)

	Comp.Ex 13	Comp.Ex 14	Comp.Ex 15
Polymer (100 parts by weight)	D	B	B
Filler (parts by weight) (particle size, $\mu\text{m}$ )	calcium carbonate 10 8	kaolin 10 12	titanium oxide 10 0.21
Stretching Ratio (times) longitudinal transverse	2.5 2.5	2.5 2.5	— —
Lubricant  (parts by weight)	montan wax  0.2	montan wax  0.2	montan wax  0.2
Stretching Ability	X	X	—
Variation Coefficient of Thickness	7	7.5	1.5
Tensile Strength (MPa) longitudinal transverse	75 70	65 64	52 52
Folding Endurance (the number of reflections)	500	600	200

### Claims

1. A stretched film of a lactic acid-based polymer comprising 3 to 25 parts by weight of one or more inorganic filler selected from the group consisting of titanium oxide having an average particle size of 0.1 to 0.5  $\mu\text{m}$ , calcium carbonate having an average particle size of 0.3 to 6  $\mu\text{m}$ , barium sulfate having an average particle size of 0.1 to 2  $\mu\text{m}$ , silica having an average particle size of 1 to 12  $\mu\text{m}$ , kaolin having an average particle size of 0.5 to 10  $\mu\text{m}$ , and talc having an average particle size of 0.1 to 10  $\mu\text{m}$  for 100 parts by weight of polylactic acid or a copolymer of lactic acid and another hydroxycarboxylic acid, and being stretched 1.3 to 5 times to one or more axial directions.
2. The stretched film of a lactic acid-based polymer according to claim 1 having an average thickness of 0.01 to 2 mm.
3. The stretched film of a lactic acid-based polymer according to claim 1 or 2 wherein the inorganic filler is titanium oxide having an average particle size of 0.1 to 0.5  $\mu\text{m}$ .
4. The stretched film of a lactic acid-based polymer according to any of the claims 1 to 3 wherein folding endurance is 900 times or more.

5. The stretched film of a lactic acid-based polymer according to any of the claims 1 to 4 wherein the film comprises 0.1 to 2 parts by weight of a lubricant for 100 parts by weight of polylactic acid or the copolymer of lactic acid and another hydroxycarboxylic acid.

5 6. The stretched film of a lactic acid-based polymer according to claim 5 wherein the inorganic filler is titanium oxide having an average particle size of 0.1 to 0.5  $\mu\text{m}$ .

7. The stretched film of a lactic acid-based polymer according to claim 5 wherein the lubricant is one or more compound selected from the group consisting of aliphatic amide-based lubricant, aliphatic ester-based lubricant and  
10 metallic soap-based lubricant.

8. The stretched film of a lactic acid-based polymer according to claim 5 wherein the film has an average thickness of 0.01 to 2mm, variation coefficient of thickness of 1.3% or less, and folding endurance of 900 times or more.

15 9. The stretched film of a lactic acid-based polymer according to claim 8 wherein the inorganic filler is titanium oxide having an average particle size of 0.1 to 0.5  $\mu\text{m}$ .

10. A use of the stretched film of a lactic acid-based polymer having an average thickness of 0.1 to 0.5mm according to anyone of claim 1 to 9, as a material of a prepaid card.  
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# EUROPEAN SEARCH REPORT

Application Number  
EP 96 11 8630

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	EP 0 510 998 A (MITSUI TOATSU CHEMICALS) 28 October 1992 * tables 3,4 * * claims 1-10 *	1-10	C08J5/18 C08K3/00
A	JP 05 070 696 A (TOPPAN PRINTING CO LTD) 23 March 1993 & DATABASE WPI Derwent Publications Ltd., London, GB; AN 93-137113 * abstract *	1-10	
A		1-10	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08J C08K
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 3 March 1997	Examiner Siemens, T
<p><b>CATEGORY OF CITED DOCUMENTS</b></p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ..... &amp; : member of the same patent family, corresponding document</p>			

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